Third United Nations International Conference on the Peaceful Uses of Atomic Energy

A/CONF.28/A/783
GERMANY, FEDERAL REPUBLIC OF
February 1964
Original: ENGLISH

Confidential until official release during Conference

Abstract

Subterraneous Disposal of Radioactive Waste in Cavities made in Salt Deposits

by

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The disposal of radioactive waste at the surface or just below surface includes the possibility of accidental or violent destruction of the stock and of dispersion of the dangerous material. Therefore, the disposal deep in the earth will have preference. Because it must be excluded that parts of radioactive matter enters the biological cycle the waste is to be protected against circulating groundwater. In most rocks - even by joints and fissures in "impermeable" rocks - groundwater is moving. No circulation takes place

1) in very great depths where groundwater - almost fossil water - stagnates also in permeable formations,

2) in few places of high mountains where a fairly dense type and a favourable position of impermeable rock can exclude any communication with the groundwater table,

3) in rocksalt (halite as rock).

Concerning liquids and gases halite is in general the most impermeable of all rocks. This fact has been proved, at many places of the world, by groundwater-, oil- and natural gas-exploration as well as by mining operations.

In Germany, experience about the practical behaviour of salt rocks has been gathered in numerous salt and potash mines during the last 80 years. For examination of the physical properties of halite, different experiments have been made in USA by the Atomic Energy Commission in Oak Ridge/Ten. in laboratories as well as in mines in Kansas. After discussions and mutual visits, German and American geologists (Bundesanstalt für Bodenforschung, Hannover, and Oak Ridge National Laboratory) came to the result that cavities in rock salt are the ideal jails for radioactive waste under any conditions. They are safe in use and cheap in arrangement.

Some provisions have to be made from the geological point of view. The halite should have a fairly considerable thickness (possibly more than 50 m) and should be pure and not have intercalations of other rocks, such as anhydrite a.o. Table-like deposits of halite which are lying almost horizontal are generally less qualified than salt accumulations in domes or diapirs. (The reasons will be explained.) The depth in which the cavities for disposal will be installed should be about 500 m or more because the natural salt flow there prevents the origin of any fissures and keeps the salt impermeable.

Salt formations where these conditions are realized are known in many places. Concerning Europe, in Germany, in the Netherlands, in Denmark, in England, also in France and Switzerland there are deposits suitable for the installation of disposal cavities. (The locations of the most suitable salt-occurrences will be reported in detail.)

To hole cavities for waste disposal two different ways can be taken. There is the possibility to use the galleries and other rooms of salt or potash mines, which were abandoned. (Details in the paper.) The operating expenses are extremely low. By drilling wells from the surface into the salt and by excavating these wells by solution of salt, rooms of about 5000 up to 200 000 m^3 can easily be obtained. The costs per m^3 are less than the expenses of a stock at the surface.

Third United Nations International Conference on the Peaceful Uses of Atomic Energy

A/CONF.28/A/784
HUNGARY
February 1964
Submitted in:
ENGLISH and RUSSIAN

Confidential until official release during Conference

Abstract

RETENTION OF FISSION PRODUCTS BY PEAT-HUMIC ACIDS, A NEW POSSIBILITY IN WASTE WATER TREATMENT

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Earlier investigations in this institute revealed that uranyl cations are strongly sorbed on insoluble humic acids and this sorption is responsible for the known geochemical association of uranium with fossil plant substances. The sorption is a cation exchange process, being effective not only for uranyl, but for many other cations of higher atomic weight.

Peat is a very cheap abundant source of the insoluble humic acids, because it consists nearly up to approximately the half of them. The behaviour of peat humic acid preparations is comparable to that of synthetic cation exchange resins of the carboxylated type.

The authors investigated the retention of fission products on peat preparations in laboratory tracer experiments. The experiments demonstrated that the majority of the fission products, the cations, are strongly fixed on peat humic acids if approximately favourable conditions are secured. The fission products can be classified according to their fixation on humic acids into the following groups:

(a) Cationic fission products. The majority of the long-living biologically dangerous fission products are cations and are firmly fixed, e.g. rare earths, Zr, Sr, Cs.

(b) Anionic fission products (I, Ru, Se, Nb, Sb, Te, etc.) and inert gases are not fixed on humic acids, not even in tracer amounts.

For an efficient binding the pH of the aqueous phase must be kept between limits of about 4-6, optimally at about 5.

Elution sequence of the long-living cationic fission products has been investigated and the so-called "breakthrough volume" determined under optimal laboratory conditions from chemically pure tracer solutions.

The application of insoluble peat humic acid for the cation exchange decontamination of medium or lower activity waste waters of the atomic industry seems to be extremely attractive. Millions of tons of peat are available in nature in the temperate and northern zones. The sorption capacity of one ton of peat is roughly about one thousand chemical equivalents. This is sufficient for the fixation of megacuries of Sr and Cs. The breakthrough volume, that is the tolerable amount of waste water, filtrable by a given quantity of peat is about 1000 - 10 000 times its quantity, if optimum conditions are secured.

Further investigations of the authors concerned the possible interference of various non-radioactive chemical contaminants which might be present in industrial waste waters. The presence of salts, complexing agents and detergents can cause displacement or even remobilisation of the sorbed fission products. The breakthrough volume is decreased resp. the necessary quantity of peat is increased by the presence of them, if their concentration is excessive.

The volume and weight of the contaminated peat humic acids can be reduced to a very small fraction by incineration before ultimate disposal.

Third United Nations International Conference on the Peaceful Uses of Atomic Energy

A/CONF.28/A/785
INDIA
February 1964
Original: ENGLISH

Confidential until official release during Conference

Abstract

A FEASIBILITY STUDY OF NON-DESTRUCTIVE ASSAY OF PLUTONIUM 239 IN IRRADIATED FUEL RODS USING SLOWING DOWN TIME SPECTROMETER

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In the region of 0.01 eV to 1.0 eV the fission cross-sections of U235 and Pu_{239} are similar except at 0.3 eV where Pu239 has a resonance peak whose value is twenty times that of U_{235}. This difference is used to determine the plutonium content by measuring the relative macroscopic cross-sections as a function of energy in this energy range. Neutrons of various energies are selected using the large slowing down time and the small energy spread of neutrons slowing down in a heavy moderator.

To test the feasibility of the method, a slowing down time spectrometer has been constructed using about 100 tons of lead (200 x 200 x 190 cms) and a low voltage pulsed accelerator using the (d,t) reaction as a neutron source.

In the experiment, pulses of fast neutrons are injected periodically into the lead assembly. The mean energy and energy spread of the slowing down neutrons are known functions of time. The fast neutrons due to fission in samples of Plutonium and Uranium are measured as a function of slowing down time and hence of energy by using a fast neutron detector with a time analyser. The neutron density at the same point is similarly measured by means of a BF_3 proportional counter.

Results so far obtained indicate that the method is feasible.

Third United Nations International Conference on the Peaceful Uses of Atomic Energy

A/CONF.28/A/786
INDIA
February 1964
Original: ENGLISH

Confidential until official release during Conference

Abstract

FUEL REPROCESSING PLANT AT TROMBAY

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A Fuel Reprocessing Plant based on the Purex process for aluminium clad uranium metallic fuel elements is nearing completion at Trombay. The Plant has provision for conversion of plutonium nitrate to any desired end product including plutonium metal. The "hot" equipment is housed in "cells" of haematite concrete walls (density 3.4 g/c.c) with thickness varying from 900 to 1,500 mm. Chemical dejacketing of the 3,400 mm. long fuel section and dissolution of uranium will be carried out in a 4,500 mm. long horizontal dissolver. Pulsed columns are used for all steps of extraction and ion exchangers are used for the final purification and concentration of plutonium nitrate. All aqueous wastes generated in the process are subjected to multiple stages of evaporation till the liquid effluents released to the environments are to drinking water tolerances. The concentrated acidic and alkaline active wastes are stored in underground reinforced concrete tanks lined with stainless steel and mild steel respectively with a total capacity of about 7,000 cubic metres.

All process equipment and piping are of AISI type 347 stainless steel. Welding for all the tanks, pipes etc. handling radioactive solutions has been carried out using inert gas arc welding. 100% of the joints have been radiographed to ASTM standards.

Exhaust air from cells and other active areas and process off-gas air are subjected to multiple stages of absolute filtration and exhausted through a 135 metres high reinforced concrete stack. Uninterrupted operation of

exhaust systems is ensured by stand-by fans and filters and provision of emergency power in case of failure of normal power supply.

When the Plant is completed, approximately 200 tonnes of stainless steel in the form of plates and structurals, 50,000 metres of stainless steel pipes, 3,000 cubic metres of heavy concrete, 20,000 cubic metres of normal concrete and 3,000 tonnes of mild steel would have been used. The plant complex with all equipment has cost about Rs. 360 lakhs (7.5 million dollars). The design, construction, fabrication of all equipment and installation were carried out by engineers of the Trombay Establishment.

The paper will indicate some of the difficulties encountered in the construction of the Plant and measures taken to overcome them.

Third United Nations International Conference on the Peaceful Uses of Atomic Energy

A/CONF.28/A/787
INDIA
February 1964
Original: ENGLISH

Confidential until official release during Conference

Abstract

CHEMISTRY OF SEPARATION OF PLUTONIUM FROM IRRADIATED FUEL

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The extraction of uranium, plutonium and mixed fission products from simulated solution of irradiated fuel by tri-n-butyl phosphate (TBP) in locally available kerosene as diluent has been studied with a view to collecting data for the design of a spent fuel reprocessing plant. The distribution coefficients for the above at different concentrations of TBP in kerosene have been determined as a function of nitric acid concentration in the aqueous phase and of the degree of saturation of the solvent phase with uranium. The data are presented in a graphical form along with the distribution coefficients for specific fission products like ruthenium and zirconium. The general trend of the results obtained, with locally available kerosene diluent, is the same as that of some other published work done under similar conditions, but differences in actual values of the distribution coefficients have been observed and the new data obtained for the distribution coefficients with respect to the degree of saturation of uranium at different concentration of TBP in the solvent phase could not be compared.

The final purification of plutonium by fixed-bed ion exchange has been investigated with three different types of resins, viz. Deacidite FF, Amberlite IRA-400 and Dowex-I x 4 with a view to establish the optimum conditions. The variation of the distribution coefficients as a function of nitric acid concentration for both uranium(VI) and plutonium(IV) in the case of Deacidite FF and Amberlite IRA-400 and distribution coefficients of solvent extracted fission products

with Dowex-I x 4 have been obtained. Absorption, break-through and elution behaviour in a column maintained at 60°C and at room temperature, have been examined. In the case of Dowex-I x 4, build-up of plutonium in the resin column due to repeated cycles of operation, has been determined and the decontamination of plutonium from fission products was found out after loading the column to three-fourth of its capacity under optimum conditions.

The mechanism of extraction of plutonium in hexone, butex and TBP has been studied by determining the distribution coefficients of plutonium by varying the nitrate ion and the hydrogen ion concentrations. From the extraction equilibria, it appears that plutonium extracts as $Pu(NO_3)_4$ from the aqueous phase into hexone and butex; but in the organic phase at higher acidity, $Pu(NO_3)_4$ changes into $H_2Pu(NO_3)_6$. Spectral changes in the organic phase in the visible range confirm these results. In TBP, plutonium nitrate is extracted as $Pu(NO_3)_4.2TBP$ which has also been confirmed by the fact that there are no spectral changes observed when organic phase acidity is varied.

The values of stability constants for nitrate complexes of plutonium(IV) at various ionic strength, determined by solvent extraction method using TBP as solvent are presented. The stability constant for sulphate complexes have also been determined using competitive method, as during reprocessing sulphamate is used for separating plutonium.

In the present process plutonium is precipitated as its oxalate from the nitrate solution and calcined to its oxide for conversion to metal through its fluoride. The decomposition of oxalates have, therefore, been studied using thermogravimetric and differential thermal analysis (DTA) methods. The kinetic parameters viz. heat of reaction, energy of activation and order of reaction have been calculated from the DTA curves.

Third United Nations International Conference on the Peaceful Uses of Atomic Energy

A/CONF.28/A/788
INDIA
February 1964
Original: ENGLISH

Confidential until official release during Conference

Abstract

HEALTH AND SAFETY PRACTICE AND MANAGEMENT IN ATOMIC ENERGY OPERATIONS IN INDIA

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Nearly a decade's experience in the control and management of radiation protection with research reactors has gone into the building up of the health and safety organization in India to meet the demands of a very comprehensive programme of large scale application of atomic energy for peaceful purposes. India has three research reactors, a fuel reprocessing plant, isotope laboratories, processing and manufacturing units for basic reactor fuels production in different parts of the country. Two high power reactors with international collaboration are to start construction soon and more are in planning. Isotopes are used on an expanding scale in the country in diverse fields.

The radiation safety and control programme in the country is based on the recommendations of the ICRP. The adoption of ICRP recommendations to national conditions involves certain difficulties of interpretation when environmental population consist of persons with diverse habits and of characteristics very different from the ICRP Standard Man. The environmental restrictions necessary to comply with ICRP recommendations for exposure of general population will have to be interpreted in the light of critical food and critical nuclides and habits of the local population. The paper presents data collected for evolving an Indian Standard Man and development of safety standards for general population as well as occupational personnel.

Selection of sites for power reactors in India take due

considerations of fission product releases from different types of reactors on site, the environmental population and utilisation of the environment in the region of 1-30 miles from site, and the acceptable site radiological burden. The paper describes national practices for siting of power reactors, planning of preoperational surveys and management of environmental problems arising out of credible accidents. The paper gives experiences of radiation exposure and control in uranium mines and thorium processing plants.

Safety management of Trombay operations are given in some detail. Hazard evaluation of reactors and processing plants are carried out and measures recommended to improve safety of operations. The paper presents experiences of normal operations as well as some unusual incidents in reactors and outlines training and emergency programmes.

Environmental monitoring surveys are carried out as a routine for presence of natural, induced or fission product radioactivity and build up of radioactivity through operational release. Environmental monitoring techniques and data as well as tracer trials carried out for study of movement of discharged effluent activity are given in the paper.

Health Physics Organisation carries out internal contamination studies of persons exposed to radiation. Whole body measurement techniques are developed for specific as well as total internal contamination. Radiochemical examination of urine is made as a routine and recommendations made to avoid over exposure of personnel. The paper presents data of human exposure for different radionuclides over years and outlines recent radiochemical methods developed for the estimation of Ra^{228}, Ra^{224}, Th^{232}, Co^{60}, I^{131}, P^{32} and other nuclides in bioassay and environmental samples.